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(54) Title: CROSS-LAMINATED MULTILAYER FILM STRUCTURES FOR USE IN THE PRODUCTION OF BANK NOTES OR THE LIKE

(57) Abstract

A laminated multilayer film substrate for use in the production of bank notes. The film substrate includes a first layer comprising at least about 50 weight percent of a high density polyethylene having a density of at least about 0.95, the first layer oriented in at least a first direction to a degree which is at least three times greater than the degree of orientation present in a direction substantially normal to the first direction, and a second layer comprising at least about 50 weight percent of a high density polyethylene having a density of at least about 0.95, the second layer oriented in at least a first direction to a degree which is at least three times greater than the degree of orientation present in a direction substantially normal to the first direction, wherein the second layer is laminated to the film substrate so that the first direction of orientation of the second layer is substantially normal to the first direction of orientation of the first layer. The resultant films having good dead-fold characteristics and other properties, making them highly suited for the production of bank notes and other security documents.

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CROSS-LAMINATED MULTILAYER FILM STRUCTURES FOR USE IN THE PRODUCTION OF BANKNOTES OR THE LIKE

The present invention relates to a multilayer film for use in the production of paper-like products such as bank notes, security documents, including travellers and bank checks, and to a process for their production. More particularly, the present invention relates to a laminated multilayer film structure having the characteristics of the high quality papers typically employed in the production of bank notes and security documents.

In the production of bank notes and security documents rag paper has been employed for over 300 years. As is well known, rag paper has several properties which are highly desirable in such applications, including dead foldability, tear resistance, printablilty and embossability.

These highly desired properties may be characterized as follows: deadfold is the ability of a substrate to be creased or folded and to retain the fold without opening. Tear resistance is the ability of a substrate to resist both initiated and uninitiated tears and punctures. Printability is the ability of the substrate to adsorb and bond inks used during the lithographic printing process. Embossability is the ability of the substrate to deform under the pressures of the intaglio printing process to form a raised image on the resulting bank note or security document, with the intaglio ink remaining on the raised, deformed region resulting in a high degree of tactility or feel to the bank note or security document. As may be appreciated, these properties combine to give bank notes and the like their familiar feel and functionality.

With the advent of color copiers and computer graphic scanners, the counterfeiting of bank notes has markedly increased. While there are active programs underway by major currency paper producers to make their substrate more secure through the use of watermarks, metallized threads and optical variable devices (OVDs), such as photochromics, holographics, and diffraction gratings these efforts do not appear to hold much promise of thwarting counterfeiters.

Plastic substrates offer a major security feature if a clear "window" is incorporated into the bank note. This window would ensure that a scanner or color copier could not copy the note. Additionally, other security features can be incorporated into or onto the bank note, including reverse printing of the note to protect the security devices and the print.

In accordance with the present invention, there is provided a laminated multilayer film substrate for use in the production of banknotes and security documents. The film substrate includes a first layer comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.95, the first layer oriented in at least a first direction to a degree which is at least three times greater than the degree of orientation present in a direction substantially normal to the first direction, and a second layer comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.95, the second layer oriented in at least a first direction to a degree which is at least three times greater than the degree of orientation present in a direction substantially normal to the first direction, wherein the second layer is laminated to the film substrate so that the first direction of orientation of the second layer is substantially normal to the first direction of orientation of the first layer.

The resultant films exhibit good dead-fold characteristics and other properties, making them highly suited for the production of bank notes and other security documents.

In forming the multilayer film substrates for use in the production of the bank notes and other security documents of the present invention, at least two layers of the substrate are to contain a major proportion of a high density polyethylene (HDPE) having a density of at least 0.95. These film layers may be composed exclusively of a single HDPE resin, a mixture of HDPE resins, or of HDPE containing a minor proportion of another polymeric material, such as low density polyethylene, linear low density polyethylene, polypropylene, ethylene vinyl alcohol (EVOH) copolymer, ethylene propylene (EP) copolymer or ethylene propylene butene-1 (EPB) copolymer, although a single HDPE resin

or a blend of HDPE resins is particularly preferred in the practice of the present invention. The use of processing additives, such as microcrystalline wax or the like may be employed with the preferred HDPE resins to yield improved processing characteristics in the extruder by reducing extruder torque. Films made with either a blend of HDPE resins or with microcrystalline wax have been found to reduce the splittiness of the film which manifests itself as the tendency of the film to break in the TD direction.

When blends of HDPE polymers are employed, such blends can comprise two or more polymers all of which preferably have densities of 0.95 or greater. Blends of HDPE polymers advantageously comprise a major proportion of HDPE having a melt index of 0.6 to 1.2 and one or more polymers having a different melt index.

Terblends may also be desirable. Suitable terblends generally comprise 50 to 98 weight percent, preferably 84 to 96 weight percent of HDPE having a density of 0.96 or higher and a melt index of greater than 0.5 to 2.0; 1 to 25 weight percent, preferably 3 to 8 weight percent of HDPE having a density of 0.96 or greater and a melt index of 0.1 to 0.5; and 1 to 25 weight percent, preferably 3 to 8 weight percent, of HDPE having a density of 0.96 or higher and a melt index of greater than 2 to 8. Preferably, the second and third HDPE polymers which are minor components are present in about equal amounts.

As will be described in more detail hereinbelow, it has been discovered that directional tear resistance in a multilayer film substrate is substantially improved when at least two of the layers of the multilayer film substrate are laminated so that the major (primary) directions of orientation for each of those layers are aligned so as to be substantially normal to one another. This improvement in tear resistance, when coupled with the excellent dead-fold, embossability and printability characteristics of HDPE resins, provides a multilayer film structure having the properties long desired in the production of banknotes and security documents.

As is particularly preferred, the film substrate of the

present invention includes a first layer comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.95, the first layer oriented in at least a first direction to a degree which is at least three times greater than the degree of orientation present in a direction substantially normal to the first direction, and a second layer also comprising at least 50 weight percent of a high density polyethylene having a density of at least 0.95, the second layer also oriented in at least a first direction to a degree which is at least three times greater than the degree of orientation present in a direction substantially normal to the first direction, the second layer being laminated to the film substrate so that the first direction of orientation of the second layer is substantially normal to the first direction of orientation of the first layer.

As may be appreciated, to achieve the object of improved tear resistance in a multilayer film of the type describe herein, it is not necessary that the first and second HDPE film layers be merely uniaxially oriented, since it has been discovered that imbalanced biaxially oriented HDPE films, laminated so that their primary directions of orientation are aligned substantially normal to each other, can perform in a substantially similar manner. A method of producing HDPE films with imbalanced biaxial orientation is disclosed in U.S. Patent No. 4,870,122.

The degree of orientation of the HDPE film layers is an important aspect of this invention inasmuch as the proper degree of orientation provides desirable physical properties. Although higher density HDPE resin having a density of 0.957 or greater can be made directly into thin films by cast extrusion, problems of curling, uniformity and flatness exist. Accordingly, thin HDPE films of 0.02 to 0.04 mm (0.8 to 1.5 mils) having the best balance of properties are obtained using imbalanced biaxially oriented films prepared from films having a cast gauge of 0.3 to 0.5 mm (12 to 20 mils) which are reduced to the desired gauge by orientation.

The films are produced and oriented in a conventional

manner. The film is heated to its orientation temperature and first subjected to MD orientation between two sets of nip rolls, the second rotating at a greater speed than the first in an amount equal to the desired draw ratio. Then the film is TD oriented by heating and subjecting it to transverse stretching in a tenter frame. Typically MD orientation is conducted at 60° to 120°C and TD orientation at 110° to 145°C.

While it is preferred that the degree of orientation in a first film direction be at least three times greater than the degree of orientation present in a direction substantially normal to the first direction, it is more particularly preferred that each HDPE film layer be oriented to an extent of 1.1 to 2.0 times in the machine direction (MD) and 6 to 12 times in the transverse direction (TD). It has been found that the HDPE film layers can be produced with excellent quality at caster speeds of up to 110 fpm corresponding to line speeds of 140 fpm at 1.25 times MD orientation.

When employed, this degree of imbalanced orientation produces an interesting effect in the HDPE components of the structure. The effect is a visible rippled and striated appearance, with the ripples being parallel to the transverse orientation direction. Under low magnification, in each square centimeter of HDPE film there will be seen from 5 to 30 discontinuous undulating ripples and striations generally parallel to the direction of orientation. This effect gives the film a slight translucent appearance, which tends to slightly blur distant objects viewed through the film. This effect indicates that the layers have been oriented in an imbalanced manner. The high density polyethylenes contemplated for use in the practice of the present invention include those disclosed in U.S. Patent No. 4,870,122.

To achieve the desired surface characteristics required of the paper-like products of the present invention, one or more skin layers can be applied, in any known manner, to the multilayer HDPE substrate material, for example by coating or coextrusion before orientation or by coating the HDPE after one or both of the orientation operations. The skin layer can be

any of the conventional material used for this purpose in conjunction with polyolefin films particularly polyethylene films. For example, to achieve a press-ready surface, a polymeric resin could be blended with fillers, fibers, pigments or the like, as necessary. Additionally, voided films, such as those disclosed in U.S. Patent Nos. 4,377,616, 4,632,869, 4,758,462 and others, may be laminated to the multilayer HDPE substrate to impart the properties of those structures to the films of the present invention.

It is also envisioned that the substrate can be embossed, dyed, printed, texturised or otherwise treated before or after lamination; this being done on the internal or external surfaces of the laminated layers, so as to provide, for example, visual and/or tactile identification of the nature of a banknote, its significance or value.

The laminating techniques which can be employed to effect the present invention are known in the art and include: adhesive-bonding or cementing, preferably with a transparent agent; solvent-bonding, where a mist of solvent is sprayed over the surfaces to be bonded together; heat-bonding where thermoplastic sheets are subject to a hot rolling or pressing operating; cast-lamination where one layer is cast onto the second and the second forms a substrate; or, extrusion or draw-lamination as in calandering operations known in the art. When optically-variable devices (OVDs) are used, they can be enclosed in pouches affixed to the substrate. On the other hand, the optically-variable devices themselves may be incorporated in one (or both) layers of the laminated substrate or between the layers, it not being necessary to incorporate a physically discrete device within a clearly defined pouch formed between the laminae.

As employed in the present specification, the term "optically-variable" is used to denote any device which can readily be made to change appearance in a reversible, predictable and reproducible manner. The appearance of such devices may be altered, for example, by the application of body-heat or manual pressure, the variation of the angle of viewing

and, the lighting conditions under which viewing takes place. The type of devices envisioned by the present invention are: diffraction gratings, liquid crystals, moire patterns and similar patterns produced by cross-gratings with or without superimposed, refractive, lenticular and transparent grids, such as Fresnel lenses, spaced partially-reflective, and partially transparent, coatings yielding variable interference patterns or the like, bi-refringent or polarising layers or zone-plates.

Generally, optically-active devices of this nature are readily recognized by unskilled persons and are yet extremely difficult to reproduce by photographic and printing techniques. Moreover the production of any one such device in a reproducible fashion and the incorporation of such a device in a plastic laminate as described by the present invention is likely to be beyond the resources of the great majority of would-be forgers. Where a flexible paper-like product such as a bank-note is sought, it is of course preferable that the optically-variable devices should, themselves, be sheet-like, flexible and thin; it is also preferable for such devices to be compatible with the plastic material employed for the laminae to facilitate bonding and mitigate against reactive changes occurring with time.

According to the present invention, one preferred form of optically variable device may be a reflecting diffraction grating consisting of a metallized thermoplastic film embossed with a diffraction pattern. To prevent access to the embossed pattern for the purpose illicit replication, it is preferable according to the present invention to employ a layer of thermoplastic material on each side of the metallized film which has similar solubility characteristics to that of the metal layer so that separation by preferential etching will be rendered extremely difficult. Another preferred device is a moire pattern formed by photographically reproducing fine line or dot patterns on each side of a thin film. The spacings of the dots and lines can be readily made too fine to be reproduced by printing techniques and yet the moire pattern can be displayed upon a much larger scale. Unique diffraction and moire patterns will often be preferred for use in bank notes and

techniques are available for producing those by computer and photo-reduction methods.

In the production of low denomination bank notes, a suitable level of security against counterfeiting may be obtained by merely a providing a clear "window" through the bank note. As indicated above, such a window would ensure that a scanner or color copier could not copy the note. Additionally, other security features can be incorporated into or onto the bank note, including reverse printing of the note to protect the security devices and the print.

The multilayer film of the present invention can also be used in applications requiring properties of high durability, and high quality printability, e.g., as labels.

The following Examples illustrate the invention; all parts are by weight unless otherwise specified.

Example 1 (Comparative)

The film of this example was produced for comparison with a film produced in accordance with the present invention.

A multilayer layer oriented film substrate having a 1.15 mil final thickness was prepared by coextruding HDPE with copolymer polypropylene skins on both sides to form a first layer (a). The HDPE resin employed was Oxychem M-6211, available from Occidental Chemical Corporation, Dallas, Texas, having a density of 0.96 and a melt index of 1.0. The copolymer polypropylene skins comprised 90% Chisso 7510, an ethylene-propylene-butene-1 terpolymer available from Chisso Corporation of Japan, and 10% Mobil LKA-753, a low density polyethylene available from Mobil Chemical Co., Norwalk, Connecticut. The HDPE comprised 90% of the resulting film layer (a) while the skins comprise 10% (5% on each side). A second film layer (b) identical to (a) was also formed. The first and second films (a) and (b) were then oriented 1.4 times in the MD at about 115°C and 6 to 12 times, e.g., 10 times, in the TD direction at 115-140°C in a tenter frame.

The copolymer skins on the inner side of oriented layers (a) and (b) were then coated with a polyethyleneimine (PEI) primer to form clear 1.15 mil OHD films.

On film (a) can be deposited optical varying devices (OVDs), at predetermined distances from one another so as to provide an OVD at the same location on each of the banknotes which are ultimately prepared from the film.

Films (a) and (b) were laminated by providing therebetween a laminating adhesive LDPE resin, Chevron 1017, available from Chevron Chemical Co., Houston, Texas. The laminating adhesive resin undergoes orienting in the machine direction during the laminating step itself, thereby imparting enhanced tear resistance in the transverse direction (TD). Lamination is carried out by conventional techniques, resulting in a multilayer film substrate having a final thickness of about 3.0 mils, the laminating adhesive resin itself providing about 0.7 mils thickness.

The resulting substrate was substantially clear and its physical properties are summarized in the Table below.

Example 2 (Comparative)

Example 1 was repeated except that the laminating adhesive resin employed was an LLDPE laminating grade adhesive resin, Dowlex 3010, available from Dow Chemical Co. of Midland, Michigan. The characteristics for the resulting substrate are set out in the Table below. Use of the higher molecular weight LLDPE as the adhesive resin in Example 2 resulted in increased TD tear strength.

Example 3

This example demonstrates that film substrates produced in accordance with the present invention possess improved resistance to tearing.

A multilayer layer oriented film substrate having a 1.15 mil final thickness was prepared by coextruding HDPE with copolymer polypropylene skins on both sides to form a first layer (a). The HDPE resin employed was Oxychem M-6211, available from Occidental Chemical Corporation, Dallas, Texas, having a density of 0.96 and a melt index of 1.0. The copolymer polypropylene skins comprised 90% Chisso 7510, an ethylene-propylene-butene-1 terpolymer available from Chisso Corporation of Japan, and 10% Mobil LKA-753, a low density polyethylene

available from Mobil Chemical Co., Norwalk, Connecticut. The HDPE comprised 90% of the resulting film layer (a) while the skins comprise 10% (5% on each side). The film (a) was then oriented 1.4 times in the MD at about 115°C and 6 to 12 times, e.g., 10 times, in the TD direction at 115-140°C in a tenter frame.

The copolymer skins on the inner side of oriented layer (a) was then coated with a polyethyleneimine (PEI) primer to form clear 1.15 mil OHD film.

A second film (layer (b)) available from Tenchi Kikai Kabushiki Kaisha, Sakai, Osaka Prefecture, Japan, prepared from HDPE resin of melt index 1-1.5, coextruded with a homopolymer polypropylene skin and stretched about 7-8 times in the machine direction, having a film thickness of 0.95 mil was laminated to layer (a) using an adhesive. The lamination was conducted so that the transverse direction of orientation of the second layer was substantially normal to the transverse direction of orientation of the first layer. The resulting laminated film had a total thickness of 2.5 mil and exhibited excellent tear strength in both the machine direction and transverse direction at 72°F.

The physical properties of the films are summarized in Table 1.

TABLE 1

		Example 1	Example 2	Example 3
Tensile	MD	6	6	-
(psi x 10^3)	TD	16	20	-
Modulus	MD	365	380	-
Stiffness	MD	17	20	-
(grams)	TD	21	24	-
Tear Strength	MD	89	63	38
(g/mil)	TD	4	5	34

CLAIMS

1. A laminated multilayer film substrate which comprises a first layer (a) and a second layer (b), which may be the same or different, each comprising at least 50 weight percent of a high density polyethylene (HDPE) having a density of at least 0.95, and each being oriented in at least a first direction to a degree which is at least three times greater than the degree of orientation present in a second direction substantially normal to said first direction; and (b) being laminated to (a) so that the first direction of orientation of (b) is substantially normal to the first direction of orientation of (a).

2. A film substrate according to claim 1 wherein for (a) and/or (b) the degree of transverse direction orientation is at least 4 times greater than the degree of machine direction orientation.

3. A film substrate according to claim 1 or 2 wherein (a) and/or (b) comprises a major proportion of a first HDPE having a density of 0.96 or higher and a melt index from 0.6 to 1.2 and a second HDPE having a density of 0.96 or higher and a melt index different from that of the first HDPE.

4. A film substrate according to any preceding claim wherein at least one of (a) or (b) is uniaxially oriented.

5. A film substrate according to any preceding claim further comprising a clear window through the bank note.

6. A film substrate according to any preceding claim further comprising, positioned between (a) and (b) is an optically-variable device (OVD).

7. A film substrate according to any preceding claim wherein the OVD comprises at least one of a diffraction grating, a liquid crystal, a moire pattern produced by cross-

gratings with or without superimposed, refractive, lenticular and transparent grids, such as Fresnel lenses, a spaced partially-reflective, and partially transparent, coating yielding variable interference patterns, a bi-refringent or polarising layer or a zone-plate.

8. A film substrate according to claim 7 wherein the OVD comprises a reflecting diffraction grating comprising a metallized thermoplastic film embossed with a diffraction pattern.

9. A film substrate according to any preceding claim in the form of a security token, such as a bank note.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US95/08042

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :B32B 3/00
US CL :428/195
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 428/195, 199, 201, 207, 211, 212, 325, 328, 332, 434, 461, 481, 483, 500, 511, 515, 518, 910

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US, A, 4,565,733 (AKAO) 21 January 1986, see entire document.	1, 2
Y	US, A, 4,865,908 (LIU ET AL) 12 September 1989, see entire document.	1, 2
Y	US, A, 3,322,613 (RASMUSSEN) 30 May 1967, see entire document.	1, 2

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

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"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
07 SEPTEMBER 1995	17 OCT 1995

Name and mailing address of the ISA/US	Authorized officer
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Form PCT/ISA/210 (second sheet)(July 1992)*

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US95/08049

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☒ Claims Nos.: 3-9
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.